Laser Diagnostics of Combustion Enhancement on a CH_4/Air Bunsen Flame by Dielectric Barrier Discharge [*]

ZHANG Shao-Hua(张少华), YU Xi-Long(余西龙)[**], CHEN Li-Hong(陈立红), ZHANG Xin-Yu(张新宇)

Institute of Mechanics, Chinese Academy of Sciences, Beijing 100190

We investigate plasma-assisted combustion for premixed CH_4/air Bunsen flames. Dielectric barrier discharge (DBD) is employed to produce non-equilibrium plasma for combustion enhancement. The transient planar laser induced fluorescence (PLIF) technique of CH and OH radicals is used to image reaction zones for enhancement measurement, and the emission spectra of the Bunsen flame are monitored to explore the kinetics mechanism. From the drift of radicals in PLIF images, the quantitative enhancement of plasma on the flame velocities of premixed methane/air flames is experimentally measured, and the data show that the flame velocities are increased by at least 15% in the presented equivalence ratio range. Furthermore, the well analyzed emission spectra of the Bunsen flame (300–800 nm) with/without DBD reveal that the emissions as well as the concentrations of the crucial radicals (like C_2, CH, OH etc.) in combustion all are intensified greatly by the discharge. In addition, the appearance of excited spectral bands of N_2 and N_2^+ during discharge indicates that the premixed gas is also heated and ionized partially by the DBD.

In modern society, it is of vital interest to maximize the efficiency of combustion to conserve fuel and to reduce pollution, in order to alleviate resource and environmental problems. However, there are many huge challenges that we first must face. For instance, achieving efficient and reliable combustion is still one of the most critical keynotes in high-speed air-breathing propulsion vehicles duo to their restricted residence time. Take the scramjet, for example, in the combustor, the flow residence time is constrained to millisecond time scales, as well as the time for chemical reaction. In this case, high flame propagation rates are highly desirable.

Researchers have shown that plasma-assisted combustion is a promising way to increase flame velocity and reduce pollutant emissions.[1−4] The reason is that plasma can produce large amounts of free electrons, ions, active radicals and excited species within combustion. These well-known crucial particles can initiate chain reactions and shift chemical equilibrium, even at low concentrations. The flame propagation speed, stability and chemical reactions are then affected and the combustion will be promoted or enhanced eventually. Because of the significant promise of plasma, a variety of plasma generator systems including dielectric barrier discharge (DBD),[5] corona discharge,[6] and microwave discharge,[7] and the nanosecond repetitively pulsed laser[8] have been used to explore plasma-assisted combustion. Most of these investigations used the flame's blow-off velocity and combustion photos to visualize the enhancement of combustion using plasma without quantitative analysis. Furthermore, the mechanisms of plasma's assistance are still under investigation.

In the present study, the plasma produced by the DBD technique is applied on a CH_4/air Bunsen flame to test and verify its enhancement for combustion. It is a preliminary step in the evaluation of this technique at laboratory scale before its application on a supersonic combustion bench test. Here the distinctive issue is that the transient PLIF images are used directly to calculate the enhancement of plasma by the imaged reaction zone. Previously, the PLIF images were generally used to demonstrate the local flame structure of combustion qualitatively.[9−11] Meanwhile, the emission spectra of the Bunsen flames were also recorded and analyzed to explore the kinetics mechanism. The DBD, also called silent discharge, is preferentially employed as a plasma source because it can be easily operated in the range from 0.1 bar to a few bars, which is the pressure range of a scramjet combustor and that of the fuel injection. In addition, it also has several other advantages such as relatively low cost in producing non-equilibrium plasma, great discharge stability, small size, easy operability of equipment, and so on.

In experiments, a coaxial configuration DBD reactor was employed because it can hide electrodes inside the burner. The structure not only makes this discharge pattern best suited to high velocity gas flow in combustion chambers, but can also combine the DBD reactor with a Bunsen combustor perfectly, just as displayed by the insert in Fig. 1. The inner electrode of the DBD reactor is a copper stick centered inside a quartz dielectric barrier tube, while a cylindrical aluminium outer electrode surrounds it. The distance between the top of outer electrode and that of the quartz tube was set as 8 mm to get a stable flame and discharge.

Figure 1 demonstrates the schematic diagram of the experimental setup. In a typical experiment, the high-voltage (HV) signals used for discharging were supplied by a homemade HV alternating cur-

[**]Corresponding author. Email: xlyu@imech.ac.cn

rent transformer and monitored by an HV probe (Tektronix P6015A) combined with a digitizing oscilloscope (Tektronix DPO4032). The premixed gas sample containing methane and air with a suitable equivalence ratio, supplied from cylinders through stainless steel pipes and controlled by an individually calibrated mass flow controller (SLA5851S Thermal Mass Flow meter, BROOKS Instrument), flows through distant pipelines and the annular space between the inner electrode and the quartz tube, where the microdischarges of the DBD form when high voltage is applied onto electrodes. The mass flow meters controlled the gas flow precisely, while the distant pipelines ensured that gases were mixed sufficiently.
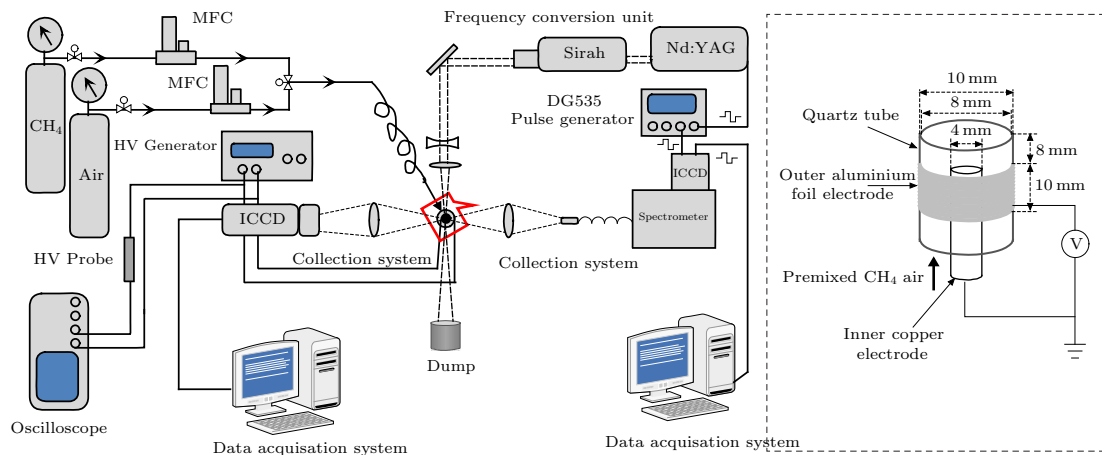

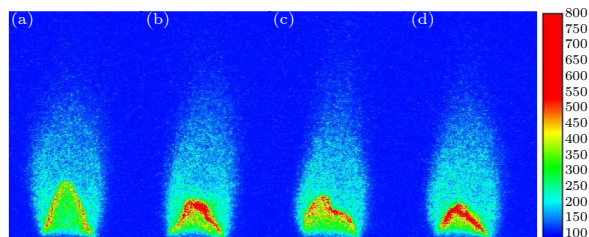**Fig. 1.** Schematic diagram of the experimental setup.

Fig. 2. CH$_4$/air Bunsen flame with (b, c, d) and without (a) DBD, the premixed gas is jetting from bottom to top.

For PLIF measurement, the fluorescence signals were emitted from excited target molecules which were created by the suitable wavelength ultraviolet laser (around 387.3 nm and 283.5 nm, respectively, for CH and OH radicals). It was generated from a tuneable Sirah laser source (Model: PRSC-GA-30, Spectra-Physics, dye: Exalite 389 for CH, Rhodamine590 and a KDP second harmonic generator for OH) pumped by the output of a Nd:YAG laser (Model: Quanta-Ray Pro250, Spectra-Physics). The output laser (the single pulse energy was set around 5 mJ, pulse width is about 10 ns) was sheeted to a laser sheet with height of 65 mm and thickness of ∼0.7 mm by a lens system before being induced into the flame. The fluorescence images were recorded by using an ICCD (Model: PI-Max, minimum optical gate width: 2 ns, Princeton Instruments) with an $f = 105$ mm UV camera lens (Nikon). The output of the ICCD was input to a digital board and then analyzed using a computer data acquisition system.

The flame emission was collected by a lens system, passed through a fiber and, then, transmitted into an echelle spectrometer (Model: Mechelle5000, spectral range 200–850 nm, spectral resolution 0.125 nm at 500 nm, Andor technology) equipped with an ICCD (Model: iStar DH734, minimum optical gate width <5 ns, Andor technology). The grating efficiency of the echelle spectrometer was calibrated using an NIST standard tungsten halogen lamp (Model: 63976, Oriel Instruments) and the wavelength calibration of the system was performed using a standard Hg-Ar lamp. Experimental synchronization was controlled through a multi-channel digital delay pulse generator (Model: DG535, Stanford Research System).

The spontaneous emission photos of a CH$_4$/air ($\Phi = 1.27$) Bunsen flame with (b, c, d) and without (a) DBD were recorded and are displayed in Fig. 2 with a long-time exposure (10 μs). The equivalence ratio (Φ) here is defined as the ratio of fuel/oxidizer (mass or number of moles) of the experiment to that of fuel/oxidizer for stoichiometric burning as follows:

$$\Phi = \frac{(\text{CH}_4/\text{O}_2)_{\text{expt}}}{(\text{CH}_4/\text{O}_2)_{\text{st}}}, \tag{1}$$

where CH$_4$/O$_2$ is the mixed ratio of fuel/oxidizer (mass or number of moles), the subscript st stands for stoichiometric conditions while the subscript expt denotes the operating conditions. The obtained photos show that the combustion has been greatly perturbed by the discharge. As shown in Fig. 2(a), without DBD, the Bunsen flame shows stable axial symmetry about the burner center, and it also demonstrates the typical Bunsen flame structure from inside to outside: inner flame, flame front (indicated by the bright curves) and outer flame. With DBD, the flame front becomes blurred and wobbly, as demonstrated in Figs. 2(b), 2(c) and 2(d). In this case, the flame front was distorted seriously when its average height was lowered obviously. It is believed that the disturbance of the flame mainly originated from the nonuniformity

of local temperature and product distribution caused by discharging. In addition, through the charged particles produced by DBD, the strong altering electric field could also impact the flame, make it wrinkled. The wrinkling and distortion would increase the flame surface and enhance the combustion even without the acceleration on combustion velocity.

Although the qualitative enhancement of DBD on combustion is displayed clearly in the emission photos in Fig. 2, the quantitative assistance of plasma could not be easily obtained because of the wrinkled and irregular flame fronts. In response to this issue, the transient PLIF technique of CH and OH radicals was employed to gain distinct flame images in our experiment. For one thing, the PLIF in nanoseconds is fast enough to "freeze" combustion to get clear and regular images instead of the wrinkled and blurred flame edges in Fig. 2. For another, previous studies have shown that the CH radical is primarily produced in the thin heat release zone of the flame (the location of flame front). Thereby it can represent the inner layer of the reaction zone[12,13] while the distribution of OH radicals can be used to determine the reaction zone[14] in combustion. Therefore, the reaction zone area, as well as flame velocity enhancement, can be extracted from the CH-PLIF and OH-PLIF images of the Bunsen flames.
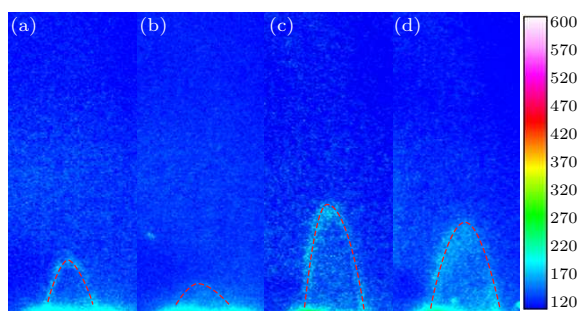

Fig. 3. CH-PLIF images of the Bunsen flame at different equivalence ratios ($\Phi = 1.27$ (a, b) and 1.38 (c, d)) with (b, d) and without (a, c) the DBD.

Two pairs of measured CH-PLIF images with and without DBD are displayed in Fig. 3 at $\Phi = 1.27$ (a, b) and 1.38 (c, d) with 10 ns exposure time. The flame fronts indicated by CH fluorescence in the figure are marked by red dashed curves for clarity. Since the position of CH radicals in the flame was apparently lowered by discharge, especially in Fig. 3(b), where CH radicals were almost fully retracted into the quartz tube by the discharge, the tangible enhancement of DBD on a Bunsen flame is exhibited more clearly by CH-PLIF images.

Furthermore, the quantitative enhancement of DBD and the flame speeds can also be obtained from the CH-PLIF images by using the Bunsen flame approach. For a laminar flame, the flame speed is defined as the velocity that a planar flame front travels toward the unburned gas in a direction normal to the flame surface.[15] Based on the discussion by Natarajan *et al.*,[16] the Bunsen flame speed can be very close to the one-dimensional reaction zone flame speed, although

the Bunsen flame has a conical shape instead of a one-dimensional surface. Therefore, the flame speed can be expressed as

$$S_u = \dot{m}/\rho_u A_b = \dot{Q}/A_b, \tag{2}$$

where the subscript b denotes the reaction zone while u means the unburned zone, S_u is the flame speed of the premixed CH_4/air Bunsen flame, $\dot{m}$ is the mass flow rate, ρ_u is the density, $\dot{Q}$ is the volumetric flow rate of the unburned mixture, and A_b is the flame area of the reaction zone. Because the location of CH radicals represents the inner layer of the reaction zone as mentioned above, the surface area measured from the CH-PLIF images can be used to replace A_b. The surface area of the CH radical area can be worked out through polynomial line fitting and spatial integral based on the axially symmetric assumption. The volumetric flow rates of CH_4 and air have been recorded separately by thermal mass flow meters. Therefore, the flame speed and its growth ratio could be calculated by Eq. (2). For accuracy and objectivity, only the growth ratio of flame speed is listed in Table 1. The data show that the flame speed increases by at least 15% in the present equivalence ratio range.

Table 1. The growth ratio of flame speed measured from the CH-PLIF images.

Equivalence ratio	Growth ratio of flame speed (Δv)
1.27	15%
1.38	19%
1.5	37%
1.63	23%
1.78	16%

A similar set of OH-PLIF images (with 5 ns exposure time) for CH_4/air Bunsen flame with and without the DBD were recorded and partly performed in Fig. 4. Images (a) and (b) came from the Bunsen flame whose equivalence ratio is 1.27. Images (c) and (d) originated from that of 1.38.
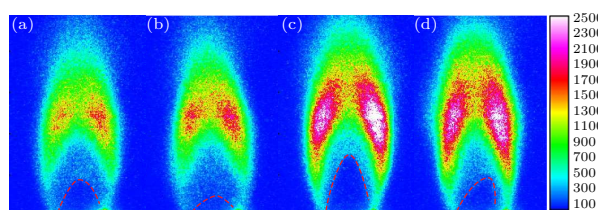

Fig. 4. OH-PLIF images of Bunsen flame at different equivalence ratios (Φ=1.27 (a, b) and 1.38 (c, d)) with (b, d) and without (a, c) the DBD.

In Fig. 4, it could be viewed that the OH-PLIF images without discharge (a, c) also show the typical Bunsen flame structure from inside to outside: inner flame, flame front and outer flame, in which the flame front is rather clear and regular as the boundary between the inner and outer flame. However, it changes with the discharge, where the clear boundary is blurred and is almost invisible as the heights of the inner flame decrease greatly. For clarity, the flame fronts in Fig. 4 are diagrammatically signed by red dashed curves. It leads to the same conclusion as

in CH PLIF study that the gas flow was accelerated by DBD, as well as the velocity of combustion.
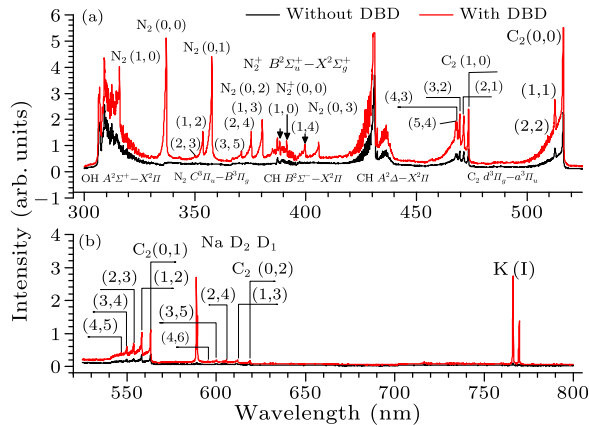

Fig. 5. The emission spectroscopy of a CH_4/air Bunsen flame with and without DBD (upper: 300–525 nm, lower 525–800 nm).

In order to determine the intrinsic flame characteristics and to study the kinetics of the active particles capable of affecting the flame speed, a detailed comparison between flame radical emission spectra with and without DBD was carried out.

As displayed in Fig. 5, the emission spectra of the CH_4/air Bunsen flame ($\Phi = 1.27$) in the range of 300–800 nm with and without the DBD were recorded, analyzed and well identified. The results show that the $A^2\Sigma^+$–$X^2\Pi(0,0)$ transition (around 308 nm) of OH radicals, the $A^2\Delta$–$X^2\Pi$ ($\sim$431 nm) and $B^2\Sigma^-$–$X^2\Pi$ ($\sim$387 nm) of CH radicals, and a series of ro-vibrational spectroscopy, like some bands in $\Delta V = 0$, -1, $+1$, $+2$ transitions (V is vibrational quantum number, the (0,0) band is located at about 516.5 nm, and the detailed assignments are labeled in the figure) of the $d^3\Sigma_g$–$a^3\Pi_u$ electronic transition of C_2 radicals, which was assigned as the $A^3\Sigma_g$–$X'^3\Pi_u$ electronic transition in some reports,[8] appearing in the flame emission spectrum. In addition, few weak atomic spectral lines were also observed, such as the double yellow lines (at 588.9 nm and 589.5 nm respectively) of sodium (Na) and the emission lines of potassium (K) at 766.3 nm and 769.7 nm. It is believed that these atomic emission lines originate from the hot quartz glass because of the combustion.

In the presence of discharge, all of the aforementioned spectral lines in flame emission were greatly strengthened, including the atomic lines of Na and K. Furthermore, besides the spectral lines mentioned above, several emission bands (the detailed assignments are labeled in the figure) of $C^3\Sigma_u$–$B^3\Pi_g$ electronic transition of N_2 and the $B^2\Sigma_u^+$–$X^2\Sigma_g^+$ electronic transition of N_2^+ appear. By integrating the peak area, the enhancements of DBD on the emission of radicals in the Bunsen flame could be acquired. For instance, the emission of OH increases by 80% (including the overlapped N_2 emission), and the spontaneous emissions of CH and C_2 radicals increase by at least 120% at their peaks. The results confirm that the

emissions of crucial radicals (like C_2, CH, and OH) in combustion are also enhanced greatly by the discharge. This means the densities of these radicals, which are major factors determining both the reaction rate and the flame speed, are highly increased by the discharge. This may explain the measured enhancement of DBD on the combustion in the CH/OH-PLIF images. In addition, the appearance of excited spectral bands of N_2 and N_2^+ during discharge shows that the premixed gases can be heated and ionized partially by DBD.

As a final note, since oxygen atoms are the main species responsible for breaking C–H bonds in hydrocarbon fuels, their emission lines have not been observed with methane in the Bunsen flame here. It is indicated that the oxygen atoms are consumed or quenched under this condition, as analyzed in previous works.[8,17,18]

In conclusion, a coaxial-cylinder dielectric barrier discharge is used to activate a premixed CH_4/air Bunsen flame. The physical appearance of the Bunsen flame and its changes are directly imaged by transient OH/CH-PLIF imaging while the emission spectra of these flames are recorded. Based on the images, the definite enhancement of the flame velocity using plasma is experimentally measured. The data show that the flame speed increases by at least 15% in the present equivalence ratio range. Furthermore, the recorded flame spectra in the range of 300–800 nm are well analyzed and identified. The results confirm that the emission intensities of crucial radicals (like C_2, CH, and OH) in combustion are also enhanced greatly by the discharge. Meanwhile, the appearance of excited spectral bands of N_2 and N_2^+ during discharge shows that the premixed gas is heated and ionized partially by DBD.

References

[1] Kim W, Do H et al 2008 *Combust. Flame* **153** 603
[2] Karpenko E I et al 2007 *Proc. Combust. Inst.* **31** 3353
[3] Starikovskii A Y 2005 *Proc. Combust. Inst.* **30** 2405
[4] Rosocha L A and Korzekwa R A 2000 *Electrical Discharges for Environmental Purposes: Fundamentals and Applications* ed van Veldhuizen E M (Huntington, NY, Nova Science) chap 10 p 245
[5] Rosocha L A et al 2004 *Phys. Plasmas* **11** 2950
[6] Wang F et al 2005 *IEEE Trans. Plasma Sci.* **33** 844
[7] Babaritskii A I et al 2004 *High Energy Chem.* **38** 407
[8] Yu X et al 2010 *Appl. Phys. Lett.* **97** 011503
[9] Tanahashi M et al 2005 *Proc. Combust. Inst.* **30** 1665
[10] Watson K A et al 2000 *Combust. Flame* **123** 252
[11] Vagelopoulos C M and Frank J H 2006 *Combust. Flame* **146** 572
[12] Sutton J A and Driscoll J F 2003 *Appl. Opt.* **42** 2819
[13] Han D and Mungal M G 2000 *Proc. Combust. Inst.* **28** 261
[14] Dally B B et al 1998 *Combust. Flame* **114** 119
[15] Law C K and Sung C J 2000 *Prog. Energy Combust. Sci.* **26** 459
[16] Natarajan J et al 2007 *Combust. Flame* **151** 104
[17] Pilla G et al 2006 *IEEE Trans. Plasma Sci.* **34** 2471
[18] Jauberteau J L et al 2002 *New J. Phys.* **4** 39